AN OFFGAS SYSTEM FOR A LOW ACTIVITY WASTE MELTER

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Abstract

The United States Department of Energy has contracted for pretreatment and vitrification of radioactive waste currently in underground storage tanks at the Hanford Site in Washington State. Waste retrieved from these tanks will be conditioned in a pretreatment facility and vitrified by heating with glass formers in up to three Joule heated melters.

Offgas generation from the vitrification process is dynamic and not steady state. The offgas consists of :

- Gases resulting from decomposition, oxidation and vaporization of feed material
- Nitrogen oxides (NOx) resulting from decomposition of nitrates and nitrites in the feed
- Chlorine, fluorine, and sulfur as acid gases, elements, and volatile salts
- Cesium as the major radionuclide
- Air in-leakage to the melter and air injected for melter operations
- Entrained particulates from feed and glass melt
- Carbon dioxide from oxidation of organics
- Small quantities of volatile organics

Before release this offgas must be treated to the point where it will meet environmental release criteria for both radioactive and chemical species. This is the main purpose of the Melter Offgas System. The major components of this system are:

Each Melter:
- Film Cooler
- Submerged Bed Scrubber
- Wet Electrostatic Precipitator

Combined Flows:
- High Efficiency Particulate Air Filters
- Exhausters
- Thermal Catalytic Oxidizer for organics
- Selective Catalytic Reducer for nitrogen oxide destruction
- Caustic Scrubber

This system maintains up to three melters under vacuum including credible surges in offgas flows. The system is largely passive with high safety margins and ready operability.

Introduction

The United States Department of Energy (DOE) and its contractors manage 177 underground radioactive waste storage tanks at the Hanford Site in Washington State. These 177 tanks contain approximately 54 million gallons of radioactive waste.[1]

It is the intention of the DOE to remove this waste from the underground tanks and provide it as feedstock to the River Protection Project – Waste Treatment Plant (RPP-WTP). The radioactive high level waste is separated into a Low Active Waste (LAW) feedstock and a High Level Waste (HLW) feedstock. The LAW feed consists primarily of the aqueous supernatant containing soluble solids presently stored in the Hanford tank system. The HLW contains the majority of the insoluble solids and radionuclides, for example technetium, transuranic elements, and cesium. This is described in more detail later.

Two vitrification systems are being designed to immobilize the radioactive waste: one for LAW feed and the second for HLW feed. Waste will be immobilized in the form of glass monoliths in metal containers. These IHLW and ILAW containers, and other containers of various secondary wastes generated during treatment operations, will be temporarily stored at the RPP-WTP and then transferred to other permitted treatment, storage, or disposal facilities.

Gases are emitted from various parts of these processes into designated treatment systems. These streams may contain radioactive particulates and radioactive and noxious gases which must be abated prior to release to the environment via a facility stack. The offgas from the LAW vitrification system is one such stream and the subject of this report.

Background

The Vitrification Process

The LAW melters convert a blended slurry of liquid LAW and glass former additives into molten glass. Sugar is also added with the glass formers to control the redox reactions in the melters. Feed is introduced to the melter and forms a "cold cap" on the surface of the molten glass before being subsumed into the melt. Up to three joule-heated ceramic melters may be concurrently operated. Under the request for proposal (RFP), the melter(s) will produce glass at a design rate of greater than 30 metric tons of glass per day.[1]

Four waste types, or "envelopes" will be treated in the RPP-WTP. These envelopes represent the maximum concentration of chemicals and radionuclides that will be found in each waste type. Only Envelopes A, B and C will be treated in the LAW melter. Envelope D waste is treated by the HLW melters, but is mentioned here for completeness and to show the path from the liquid portion of the Envelope D waste to the LAW melter.

- Envelope A. This LAW feed envelope will contain cesium and technetium at concentrations high enough to warrant removal of these radionuclides during pretreatment in order to ensure the immobilized LAW glass meets applicable requirements.
- Envelope B. This LAW feed envelope will contain higher concentrations of cesium than Envelope A. Both cesium and technetium must be removed to comply with the contract specifications for ILAW. This envelope will also contain concentrations of chlorine, chromium, fluorine, phosphates, or sulfates, that are higher than those found in Envelope A, and which may limit the waste loading in the glass.
- Envelope C. This LAW feed envelope will contain organically complexed strontium and transuranic (Sr/TRU) compounds that will require removal in a processing step unique to this waste envelope. As with Envelopes A and B, cesium and technetium will also require removal in the pretreatment process to ensure that the contract ILAW specifications can be met.
- Envelope D. HLW feed envelope will be in the form of a slurry containing 10-200 grams of solids per liter of slurry. The liquid fraction of the slurry will be comprised of Envelope A, B, or C waste, and the solid fraction will be Envelope D waste.

Offgas Design Parameters

Offgas is generated from the vitrification of LAW. The rate of generation of gases in the melter is dynamic and not steady state. The LAW melter evolves steam and gases resulting from decomposition, oxidation and vaporization of feed material. Additional air or steam may be used to agitate the molten glass and control the melt rate and redox reactions. There is also air in-leakage since the melter is kept under a vacuum to control emissions. These gases may entrain material from the cold cap or the molten glass and carry it out of the melter.

The melter offgas consists of:

- Gases resulting from decomposition, oxidation and vaporization of feed material.
- Nitrogen oxides (NO_x) from decomposition of metal nitrates in the melter feed.
- Chloride, fluoride, and sulfur as oxides, acid gases and salts.
- Cesium, strontium, technetium and transuranic elements as the major radionuclides.
- Air in-leakage to the melter caused by operating the melter under vacuum.
- Air used intentionally to control melter operating parameters
- Entrained particulates from dried feed material and molten glass.

In addition, the LAW melters release small quantities of semi-volatile and volatile organic compounds and radioactive gases. These radionuclides include elemental iodine (^{129}I), carbon (^{14}C) as carbon dioxide, and tritium (^{3}H) as water. The semi-volatile and volatile organic compounds consist of degradation products of organics in the feed as well as degradation products from sugar.

Limited quantification of offgas parameters has been done through prototype and bench scale testing with simulants. Additional work will be performed to further validate mass and heat balances and assign operational constraints.

Previous melter experience has shown that surges (temporary flowrate increases into the melter offgas system) are a regular occurrence in melter operation. Therefore, in addition to normal offgas flows, the LAW offgas treatment system is designed to treat intermittent surges of up to seven times the normal rate for condensable gases, such as steam, and up to three times the flow rate for non-condensable gases.

Previous Melter Offgas Experience

Radioactive and simulated radioactive waste has been vitrified in a variety of melters from bench scale and prototypes to production units both in the United States and abroad. Offgas systems used on these other melters were evaluated. As addressed in the last section, valuable information on surges is available from previous melter offgas experience. Other information may be ascertained by evaluating the similarities and differences between challenges and equipment used in other melter offgas systems versus the LAW melter offgas system. Most prominently explored were the systems operated at the Defense Waste Processing Facility (DWPF) on the Savannah River Site, SC, at the West Valley Demonstration Project (WVDP) in West Valley, NY, and previous GTS Duratek melter projects around the DOE complex.

Two production melters, DWPF and WVDP, have been operated to vitrify high level radioactive waste. In addition, several laboratory and pilot scale units have been operated for both simulants and actual radioactive waste. Similar to all melter offgas systems are the challenges of treating high temperature gases and general vitrification products. Melter offgas is thermally hot with temperatures ranging from 300 °C up to 1000 °C. Vitrification products, from simulated or radioactive feed, include gross entrainment of feed material, gases from decomposition of feed components, volatized material, and glass particles. Each of these products, if from radioactive feed, will contain radioactive material.

Melter offgas can be treated by dry or wet processes. The main advantages of dry processes are that the offgas stream does not require cooling and there is no secondary aqueous stream generated, however dry processes have not been used in nuclear applications except for laboratory scale units. Therefore the first challenge in treating melter offgas is to cool the offgas to the point where aqueous processes can be used. Particles and any gaseous contaminants are then removed to acceptable levels prior to release to the environment.

For cooling the melter offgas, DWPF uses an ejector venturi scrubber while WVDP uses a submerged bed scrubber. In addition to reducing the temperature of the melter offgas, both units also remove the bulk of the larger diameter particles as well as some of the gases (primarily water-soluble gases). The next unit operations are intended for submicron particulate removal. DWPF uses a steam atomized scrubber followed by a High Efficiency Mist Eliminator (HEME) while WVDP uses only a HEME. In each system these devices are followed by High Efficiency Particulate Air (HEPA) filters. Two HEPA filters in series provide the bulk of the particulate removal, with a minimum decontamination factor of 100,000. The equipment upstream of the HEPA filters provides a joint decontamination factor of from several hundred to several thousand. On a weight basis, however, the devices upstream remove a larger amount of contaminants and may be thought of as extending the life of the HEPAs to a practical level by preventing them from loading too rapidly.

Removal of different constituents by the various melter offgas systems is mediated by different feed sources, different feed preparation steps, and different regulatory requirements. DWPF does not need devices to specifically remove radioactive or noxious gases in the melter offgas as these are removed by condensation. West Valley must reduce NO_x emissions and uses a selective catalytic reduction unit.

Offgas Equipment And System Selection

Systems Engineering Approach Overview

Offgas Systems have developed from single unit operation remediation techniques to integrated systems to treat multiple contaminants. From an article on *Controlling Emissions From Fuel and Waste Combustion*, "the traditional approach to emissions control for combustion fluegases is to use a series of control systems in succession, each targeting a different pollutant . . . such a treatment train is both complex to operate and expensive". The alternative then is to develop "'multiple pollutant' control systems, which can adequately destroy or neutralize a variety of pollutants in a single unit".[2]

Choosing equipment trains for contaminant removal requires a holistic approach to contaminant remediation. As each of the control technologies has advantages and disadvantages in specific applications, the controls considered must be evaluated in the environment under which contaminants will be emitted. Some control technologies work most efficiently in combination with other pieces of equipment. Some of the equipment is chosen for its ability to perform dual purposes. In addition, total control of a specific contaminant should be evaluated over the entire facility. For LAW this includes the pretreatment processing prior to receiving the feed into the LAW facility. Processes for removing components in the feed (i.e. cesium and technetium) may remove or add other components as well.

A systems engineering approach was used to select the appropriate equipment and equipment train to satisfy the requirements. The building blocks to construct an adequate system consist of identifying functions and requirements, selecting alternatives that meet the requirements, developing criteria for making a selection from the alternatives and then nominating a preferred alternative based on degree of conformance with the criteria.

Functions and Requirements of the Melter Offgas System

The offgas system must perform the following functions:

- Cool the melter exhaust
- Remove larger particulates and aerosols (>1 μm)
- Provide high efficiency removal of sub-micron particulates
- Destroy or remove organics
- Control NO_x emissions
- Remove acid gases
- Provide a pressure confinement boundary

In designing an offgas system, once the basic functions of cooling, noxious gas destruction or removal, and solids removal have been addressed, the next emphasis is on improving the reliability, safety, maintainability and operability of equipment. Access to offgas equipment is limited, and may not be possible because of radiation levels. In addition the consequences of failure to perform may be large in terms of hazards to personnel or the environment and in costs to recover. To meet all these needs, devices that are passive with little or no maintenance requirements, requiring limited operational attention, and with slow and readily detected failure modes are desirable.

It is the purpose of the offgas system to cool and treat the melter offgas to the point where it will meet release criteria for radioactive and chemical material. In addition, the requirement for each function is that the equipment chosen must be the best available equipment or system of equipment to perform that function. The Best Available Radionuclide Control Technology (BARCT) and Best Available Control Technology (BACT) processes were used to assure that the equipment chosen was consistent with the air permit application requirements

Other requirements must be met with respect to flowrate requirements and pressure control. There are number of fixed air in-bleeds and six steam or steam and air in-bleeds into the LAW Offgas System (this includes all three melters). This supports functions including redox control, agitation, pressure control, and film cooler. The offgas system must also provide a pressure confinement boundary that will control melter pressure and prevent vapor release to the cell, which would increase contamination levels and could exceed release limits or cause structural damage to the melter and associated components.

Alternatives Evaluation Overview

For each function required, various pieces of equipment were selected and evaluated. Hazards related to dose rates, releases, and exposure for each equipment item were identified. Once the equipment items were identified to meet the performance requirements they were configured into offgas system alternatives and the systems were evaluated using identified weighted criteria. The sequencing of treatment equipment was a major factor for developing systems since sequence is important to operating efficiency, and thereby influences equipment sizing.

Equipment items were evaluated on the basis of relative performance, reliability, safety, remotability and space requirements. Other aspects taken into consideration were utilities required, maintenance requirements, ease of operation and effects on related equipment or systems.

Evaluation of Equipment and Systems

A complete alternative evaluation was conducted and a summary of the equipment evaluated per function and the chosen melter offgas system is included here.

Offgas Cooling and Removal of Larger Aerosols

Two major pieces of equipment were considered for the quenching of the offgas and removal of larger particulates, an ejector-venturi scrubber (EVS) and a submerged bed scrubber (SBS). By using wet processes, the water used comes in contact with the contaminated air stream resulting in a contaminated water stream. Minimizing the amount of contaminated water reduces disposal costs.

Other cooling and large particle removal equipment considered include:

- Cyclonic Scrubbers
- Hydrosonic Scrubber (HSS) – Air-atomized Liquid or Steam
- Impingement-Plate/ Tray-Tower Scrubber
- Mechanically-Aided Scrubber
- Packed-Bed/Packed-Tower Wet Scrubber
- Spray-Chamber/ Spray-Tower Wet Scrubber

High Efficiency Particulate and Aerosol Removal
Five major pieces of equipment were considered for this melter offgas function: High Efficiency Mist Eliminator (HEME), HEPA filters, Wet Electrostatic Precipitator (WESP), Hydrosonic Scrubber (HSS) and High Efficiency Metal Filters (HEMF).

Other particulate and aerosol removal equipment considered include:

- Dry Electrostatic Precipitator (ESP)
- Baghouse (Fabric Filter)
- Deep Bed Glass Fiber Filter
- Deep Bed Sand Filter
- Prefilter
- Ultra Low Penetration Air (ULPA)/Very large Scale Integration (VLSI) Filter
- Standard Mist Eliminator

Volatile Organic Compounds (VOC) Destruction
Three techniques were evaluated for this offgas system requirement: granular activated carbon bed, selective catalytic oxidation, and thermal oxidation. All have been used in a nuclear setting.

Control of NO_x Emissions
Three equipment items were considered for this offgas requirement: Selective Catalytic Reduction (SCR) with ammonia, $SCONO_x$ Catalytic Absorption System, and a Wet Caustic Scrubber. All these equipment items have had extensive use industrially.

Other NO_x removal equipment considered included Selective Non-Catalytic Reduction (SNCR)

Acid Gas/Iodine Removal
Three techniques were evaluated for acid gas removal: Caustic Scrubber, Calcium Hydroxide $Ca(OH)_2$ Feed Addition, and Wet Lime Scrubbing. It is uncertain if enough elemental iodine will be present to warrant an additional system to remove iodine from the offgas stream. At this point it is not required; however if a separate system is needed, three major pieces of equipment will be considered: Silver Based Zeolite Bed, Silver Based Mordenite Bed and Silver Nitrate Loaded Silica Gel.

Other acid gas/iodine removal equipment considered include:

- Silver Nitrate Scrubber (Silver Reactor)
- Activated Carbon Bed Adsorber
- Iodox Process
- Mercurex Process

The following table provides a summary of the evaluated feasible equipment/techniques and the removal efficiency associated with them.

Table 1 - Equipment vs. Contaminant Matrix

Function/ Unit Operation/ Equipment	Material Type			
	<1µm	>1 µm	Radioactive Gas	Permitted Gas
Cooling of Melter Exhaust and Removal of Larger Aerosols				
Ejector Venturi Scrubber	F	E, DF=10-50	P	F
Submerged Bed Scrubber	F	E	P	F
Steam Film Cooler	N/A	N/A	N/A	N/A
Air Film Cooler	N/A	N/A	N/A	N/A
Direct Liquid Contact Condenser	P	F	F, Condenses ^3H	F
High Efficiency Aerosol Removal				
High Efficiency Mist Eliminator	G	E	P	F
HEPA Filters	E,99.97% Eff.	E	N/A	N/A
Wet Electrostatic Precipitator	E	E	P	F
Hydrosonic Scrubber	E	E	P	F
High Efficiency Metal Filter	E	E	N/A	N/A
Control of NO_x Emission				
NO_x SCR	*	*	N/A	E,99.7% Eff.
$SCONO_x$	*	*	N/A	E
Wet Caustic Scrubber Column	P	F	G	G,60% Eff.
Iodine Removal				
Silver Based Zeolite Bed	*	*	DF=10^2 to 10^5	N/A
Silver Based Mordenite Bed	*	*	DF=10^3 to 10^4	N/A
Silver Nitrate Loaded Silica Gel	*	*	DF=10^4	N/A
Organic Destruction				
Granular Activated Carbon Bed	*	*	P	G, 95% Eff.
Organic SCO	*	*	N/A	E

DF = Decontamination Factor (estimate, if available)
If quantitative information is not available, a qualitative assessment will be used.

E = Excellent	P = Poor
G = Good	N/A = Not Applicable
F = Fair	* = Can cause plugging

Evaluation of Chosen Melter Offgas System

All the alternatives assumed a film cooler would be used. Offgas from the melter is vented to the melter offgas system through a film cooler. The main purpose of the film cooler is to cool the offgas sufficiently to minimize the deposition of solids on the pipe wall. Downstream of the film cooler offgas train alternatives were evaluated using the following criteria.

- Low or no maintenance - The main objective of this criterion is equipment that requires low or no maintenance. A low maintenance equipment item is one that requires minimal or no routine attention to maintain function throughout its design life. High maintenance equipment would require more frequent adjustment or servicing.
- Functional for long term operation - Equipment characterized with high life expectancy means that it would last more than one melter cycle (3 to 5 years). One with moderate life is replaced as often as the melter. One with low expectancy is replaced more often than the melters.
- High Reliability - Highly reliable equipment meet performance requirements with low risk of failing in service. Low reliable equipment would be one prone to failure more often.
- Passive/no moving parts - Equipment is considered passive when it has no moving parts. For example, a piece of equipment is considered passive if it does not require a mechanical pump to operate.
- Space efficient - Space efficiency depends on how much equipment is needed and how large the equipment needs to be to perform the required duty. It also takes into account location of the equipment i.e. the amount of shielded space required.
- Slow and readily detected failure modes - Equipment with slow and readily detected failure modes gives a warning before equipment failure occurs so that an effective response is possible.
- High efficiency - High efficiency equipment is that which performs its task very effectively without the need for extra stages or other equipment.
- Low Technical Risk - Low technical risk equipment has demonstrated effective performance in similar applications. New technology or equipment that has no track record in the type of application sought is considered high risk. Limited experience or novel applications of demonstrated technology is considered moderate risk.
- Easy to operate - Easily operable equipment is simple and does not require extensive process monitoring or intervention for operation.
- Require a minimum of human intervention - An equipment requiring low human intervention is one that is highly reliable, passive, requires low maintenance and has low failure rate.
- Tolerant to a wide range of process (operating) parameters - Equipment able to perform the required task successfully during normal, turndown, and transient conditions is said to be tolerant to a wide range of process parameters.

There are a number of regulatory and other measures meant to provide design guidance beyond readily apparent needs such as low cost and minimal technical risk. Specific criteria would be 1) minimal waste discharged to the environment 2) lowest integrated exposure of plant personnel to ionizing radiation 3) effective control technology without undue cost 4) least secondary waste including spent or failed equipment.

LAW Vitrification Off-gas Treatment

Primary LAW Melter Offgas Treatment System Overview

The primary melter offgas system cools the offgas and removes the bulk of the particulates and aerosols generated by each melter. Each of the three LAW melters vents to their own primary offgas treatment system, consisting of the following components:

- Film cooler (using air or a mixture of steam and air)
- Submerged Bed Scrubber
- Wet electrostatic precipitator (WESP)

The film cooler reduces the temperature of the offgas before it enters the SBS, where offgas is drawn through a flooded packed bed. There, steam is condensed, the gas further cooled, and larger particles, aerosols, and soluble solids removed. The WESP provides high-efficiency removal of sub-micron particulates and aerosols, and protects the downstream HEPA filters from the need for frequent replacement. Ionizing electrodes in the center of tubes within the WESP create a strong electrical field that negatively charges particulate matter entrained in the offgas stream. The charged particles are removed from the offgas as they adhere to the positively charged tube walls.

Secondary LAW Offgas Treatment System Overview

The offgas ventilation ducts from the three primary offgas treatment systems are combined with the LAW vessel ventilation header before entering the secondary LAW offgas treatment system. The secondary offgas treatment system consists of the following components:

- Heater/HEPA -filtration unit
- Thermal catalytic oxidation unit
- Selective catalytic reduction unit
- Caustic scrubber

The HEPA filters complete removal of the vast majority of particulates and aerosols. The thermal catalytic oxidizer removes VOCs from the offgas stream, after which the catalytic reducers remove nitrogen oxide using ammonia as a reducing agent. The offgas stream then enters the packed caustic scrubber column, moving countercurrent to a trickling caustic liquid stream. This removes iodine-129 as well as sulfur dioxide. After passing through this treatment train, the offgas is released to the atmosphere through the LAW vitrification plant stack.

Within the LAW vitrification facility, waste tanks are kept under a slight vacuum to control emissions. Vessel ventilation lines are connected to the main vessel ventilation header, which joins the primary offgas treatment system lines downstream from the WESP and immediately upstream from the preheater for the secondary offgas system HEPA filters. The combined melter offgas streams and the vessel ventilation offgas stream then goes into the LAW secondary offgas treatment system described above.

Figure 1 provides a block diagram indicating the placement of each piece of equipment in the melter offgas treatment train.

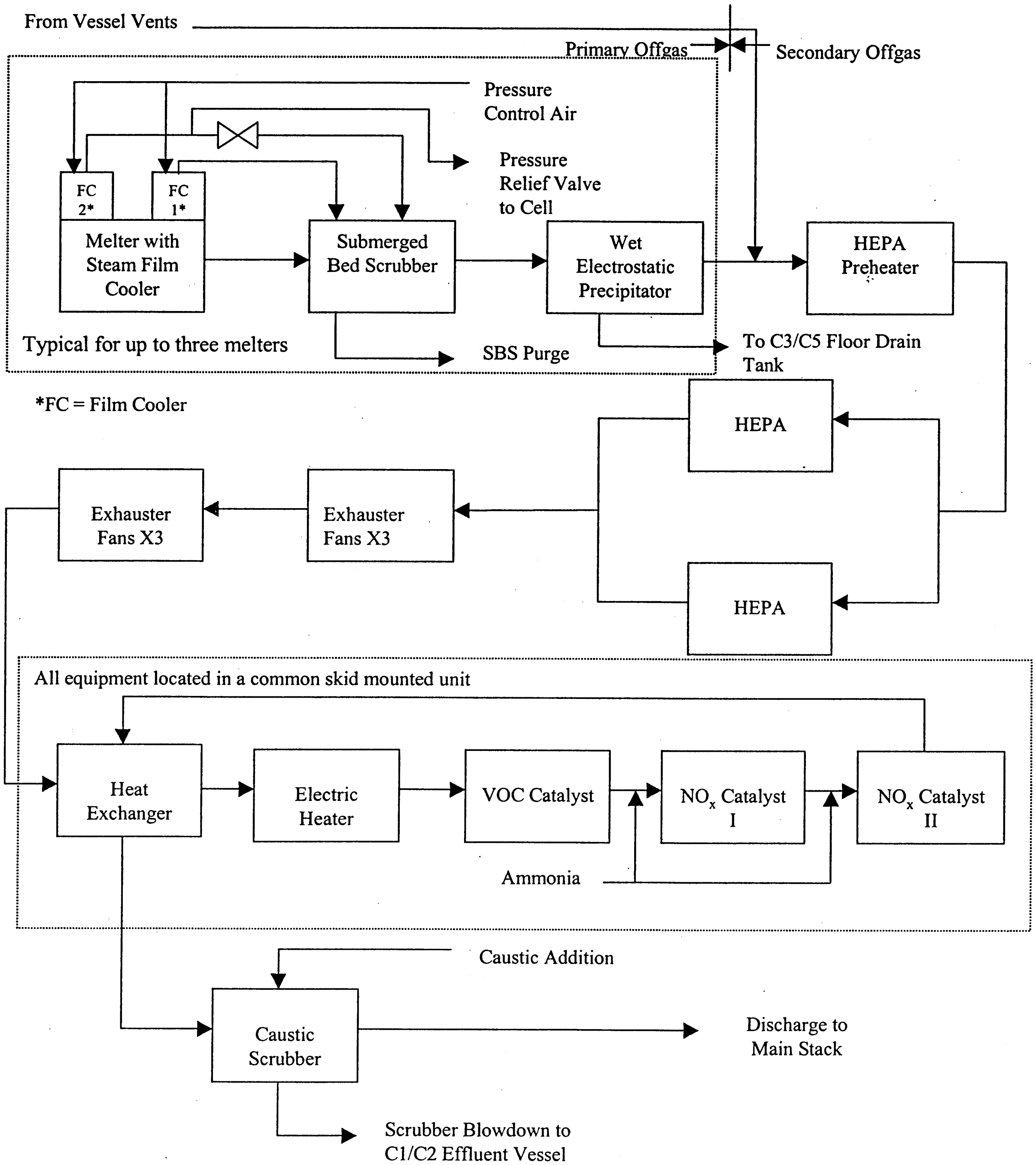

Figure 1: LAW Melter Offgas Treatment System Block Diagram

Melter Offgas Pressure Control

Pressure Control of Multiple Melters

Melter vacuum is maintained in several ways:

- Long term changes, such as increases in air in-leakage as melters age, are met by changing the exhausters' speed.
- Short-term changes such as variations brought about by the non-steady state regime in the melter are met by adjusting air/steam flow to the film cooler and the amount of melter pressure control air.
- When the melter vacuum decreases to a set point, the standby offgas line is activated to prevent melter pressurization.
- In case of a surge above that to which the melter offgas system and the standby line are designed for occurs, a pressure relief valve provides melter pressure relief.
- The line between the melter and the SBS is sized to accommodate the design basis condensable surge without causing the melter vacuum to decline to the point where there is a risk of pressurization. Condensable surges are quenched in the SBS and do not significantly affect downstream operations. Non-condensable surges are accommodated by reducing the melter pressure control air and by the decline in in-leakage that occurs with reduced melter vacuum.
- The underlying design premise is that non-condensable flow from each melter is constant so that surges in one melter do not cause pressure changes downstream of the SBS and hence do not propagate to other melters.

Off-Normal Pressure Control

Infrequently, each melter may experience a reduction in vacuum, which will actuate an alternate route into the offgas treatment system. The following are abnormal situations that would require diversion:

- Plugging of the air film cooler line from the melter to the SBS
- Melter surge in excess of the melter offgas system design basis of seven times the volume of condensable gases and three times the volume of non-condensable gases

Automated gas pressure monitoring, alarms and control devices will detect these conditions. Each LAW melter will have an alternate route into the offgas treatment system. The alternate route will consist of a fast-actuating butterfly valve and a redundant bypass film cooler line from the melter to the SBS.

In the event of a plug developing in the line from the melter to the SBS, or a surge in offgas resulting in reduction of vacuum below the minimum normal set point, the butterfly valve will open the alternate route to the SBS. Air and steam will flow through the bypass film cooler to cool the offgas and minimize the deposition of solids on the pipe wall.

Detailed Equipment Description

Steam/Air Film Cooler

The offgas exits each melter between 300 and 500°C and is mixed with steam or a steam/air mixture in the Offgas Film Cooler. The primary function of the film cooler is to cool the offgas to minimize solids deposition on the offgas piping walls. The film cooler is a double-walled pipe designed to introduce air/steam axially along the walls of the offgas pipe through a series of holes or slots in the inner wall. The steam or steam/air flow along the pipe wall mixes with and cools the offgas. This stream injection helps maintain an offgas flow velocity of around 50 ft/s. This velocity helps minimize solids deposition within the film cooler and offgas line.

Additional process air (beyond what is needed to cool and accelerate the melter offgas) is also introduced downstream of the film cooler to facilitate melter plenum pressure control. The flowrate of this air is controlled via a pressure control valve so that a steady melter pressure is maintained, even as the melter offgas generation rate fluctuates. The addition of pressure control air further cools the offgas.

Because the offgas line from the film cooler to the SBS is prone to solids deposits, means of cleaning this line are necessary. Three methods have been selected at this point. The first method is a water spray into the film cooler and the offgas line to dissolve the solids. If this method proves to be ineffective, a mechanical reamer may be used. A flange in the film cooler is provided for the reamer insertion. If the mechanical reamer fails to clean the film cooler and the offgas line, then they may be replaced.

Offgas Line from the Melter to the SBS
A 10 inch diameter sloped line connects the film cooler to the SBS. This line is designed to handle surges up to seven times (7X) steam and three times (3X) non-condensables without causing melter pressurization. This line rises from the melter and then slopes downward towards the SBS.

This line is the major potential failure point in the offgas system due to solids deposition. To prevent pressurization of the melter and to provide more flexibility to the main offgas system, a standby line is provided that is nearly identical except for the addition of a butterfly valve as the isolation device.

To maintain the line clean and prevent plugging, a small back flow of air is allowed through the line. Means of line cleaning in case of plugging are identical to the ones for the main offgas line. In the event that the melter surge exceeds the melter pressure limit, a pressure relief valve opens allowing melter venting to the process cell. The process cell is not normally occupied and vents through a high capacity HVAC system which includes HEPA filters.

Submerged Bed Scrubber (SBS)
After the film cooler the offgas enters the Submerged Bed Scrubber (SBS) for further cooling and solids removal. The SBS Column Vessel has a capacity of 3,000 gallons. The SBS is a passive device for aqueous scrubbing of entrained radioactive particulate from melter offgas, cooling and condensation of melter vapor emissions, and interim storage of condensed fluids.

The SBS has two offgas inlets, one for the normal operations line and one for the standby line. The offgas enters the SBS through the inlet pipe that runs down through the bed to the packing support plate. The bed retaining walls extend below the support plate creating a lower skirt to prevent gas bypassing the packing. A hold-down screen is used to prevent the bed from being carried out at high gas velocities. Gas bubbles form and rise through the packed bed. The reduced density causes the liquid to circulate up through the packing, and hence downward in the annular space outside the packed bed. The liquid circulation helps to prevent buildup of captured material in the bed by constantly washing the material away.

A cooling jacket located on the scrubber vessel and cooling coils inside the vessel maintain the scrubbing liquid at 50°C. Recirculation of cooled condensate from the SBS Condensate Vessel also contributes to the SBS Column Vessel cooling. The cooling medium for both vessels is chilled water. As the offgas cools, water vapor condenses and increases the liquid inventory. Water overflows continuously into the SBS Condensate Vessel thereby maintaining a constant liquid depth in the scrubber.

The scrubbed offgas discharges through the top of the SBS and is routed to the Wet Electrostatic Precipitator (WESP) for further particulate removal.

Solids captured in the SBS are maintained in suspension via a recirculation pump located in the SBS Condensate Vessel that recirculates the condensate to the SBS Column Vessel. This pump has a dipped leg discharge into the SBS Column Vessel that agitates the bottom of the vessel.

Condensate produced and solids captured in the SBS Column Vessel are purged periodically. The Scrubber Water Purge Pump in the SBS Column Vessel transfers the condensate to the SBS Condensate Collection Vessel.

Wet Electrostatic Precipitator (WESP)

After removing larger particulates and aerosols in the SBS, the cooled offgas at 50°C is routed to the Wet Electrostatic Precipitator for high efficiency removal of sub-micron particulates and aerosols. The offgas enters at the top of the unit and passes through a distribution plate. The saturated gas then flows downward through the tubes of the WESP.

The tubes act as positive electrodes. Each of the tubes also has a single negatively charged electrode, which runs down the center of the tube. A high voltage transformer/rectifier supplies the power to these electrodes. A strong electric field is generated along the electrode giving a negative charge to particles as they pass through the tubes. The negatively charged particles move towards the positively charged tube walls and are intercepted. As the gas passes through the tubes, some of the first particles captured are the water droplets. As the water droplets gravity drain down the electrode tubes the collected particles are washed off and the rundown is collected in the WESP dished bottom area. The collected condensate then gravity drains into the Effluent Vessel. Each WESP is equipped with a spray wash ring for periodic washdown and maintenance.

The WESP transformer/rectifier is located outside of the process cell. This allows for hands-on maintenance to the transformer if required. Process air is added through the electrode ducting to keep the electrode dry.

Downstream of the WESPs the individual offgas lines combine with the flow from the vessel vent header.

Secondary Offgas Treatment System

The combined primary melter offgas streams and the vessel ventilation offgas stream are submitted for treatment through the secondary offgas treatment system. This system removes almost all remaining particulate, miscellaneous acid gases, NO_x, and volatile organic compounds.

HEPA Preheaters, Filters and Exhauster

The offgas is subjected to further particulate removal with HEPA filters. The combined offgas stream is passed through the Offgas HEPA Preheater. This electric heater increases the gas temperature 10 to 20°C above its dew point to avoid condensation in the HEPA Filters. Two electric heaters in series, in a common housing, are provided. One is operational while the other one is on standby mode.

The offgas then passes through HEPA filters. Design capacity for this unit is approximately 8,000 CFM. To obtain 99.999% removal efficiency two sets of filters are located in series. There are two parallel sets of HEPA filter trains. The offgas passes through one train while the other remains available as an installed backup. When the differential pressure across the filters or the radiation level reaches set points, they are changed out. Filter banks are contact maintained and the filters are changed one at a time. The location of these HEPA filters also protects the downstream equipment from radioactive contamination.

Three variable speed exhauster sets, the Offgas Exhauster Fans are located downstream of the HEPAs. Each exhauster set is rated at 50% of the total system capacity. One or two sets of exhausters will be working at the same time, depending on how many melters are operating. At this point the offgas is approximately 84°C and 25% relative humidity.

Catalytic Oxidizer/Reducer Unit

The offgas has a high level of gaseous nitrogen oxides (NO_x), since the melter decomposes the parent nitrate/nitrite compounds. Volatile Organic Compounds (VOC) are also present in the offgas stream and require removal to meet regulatory limits.

To meet these requirements, the offgas is passed through a catalytic oxidizer/reducer unit. This skid mounted unit houses the Catalytic Oxidizer Heat Recovery Unit the Catalytic Oxidizer Electric Heater the Catalytic Oxidizer VOC catalyst and the Catalytic Oxidizer SCR Catalysts 1 and 2.

As the gas enters the unit it passes through a plate heat exchanger for waste heat recovery. This heat exchanger raises the offgas temperature to about 350°C. The heating medium used is the exhaust from the catalytic oxidizer/reducer unit at approximately 515°F. The offgas then passes through an electric heater which is operated as necessary to bring the temperature up to between 350°C and 400 °C for the VOC catalyst to operate. The heater is mainly used during startup and low NO_x concentration. During times of low NO_x concentrations the catalyst columns will not heat the offgas enough to heat the incoming offgas.

The VOC catalyst column operates at a lower temperature (minimum 350°C) than the NO_x catalyst (minimum 400°C) therefore it is placed at the beginning of the unit. The VOC catalyst is a platinum-based material deposited on a metal monolith, which is held in frames and inserted and removed through access doors. The VOC destruction reaction is exothermic. Through this catalyst the organics are decomposed to carbon dioxide, water vapor and possibly acid gases (depending on any halogenated VOC present in the stream).

After the VOC catalyst, the offgas enters a chamber where a gas mixture of ammonia, CO_2 and water vapor, is injected through an atomized spray and allowed to mix with the offgas. Ammonia is added so that it can react with the NO_x and reduce it to nitrogen and water vapor. The major reactions occurring at the catalyst are:

$$4NO + 4NH_3 \rightarrow 4N_2 + 6H_2O \qquad (1)$$

$$2NO_2 + 4NH_3 \rightarrow 3N_2 + 6H_2O \qquad (2)$$

At this point the offgas temperature is around 400°C, well above the formation temperature of ammonium nitrate (it decomposes above 210°C). The offgas then goes through the first set of NO_x catalyst modules. Since the amount of NO_x present in the offgas is high enough and the required removal efficiency is >98%, two sets of NO_x catalyst columns are used. After the offgas passes through the first set, more ammonia gas mixture is injected and allowed to mix and then sent through the second set. The NO_x catalyst is a titanium oxide based material deposited on a metal monolith, which is held in frames and inserted and removed through access doors.

The ammonia slip exiting this system is limited to 50ppm. Instrumentation is located at the outlet of the unit to determine the ammonia concentration, NO_x levels and VOC levels. Ammonia will be produced on-site as needed. This is done by dissolving urea pellets in de-ionized water to a concentration of 40% by weight and then heating the solution in a pressurized hydrolysis reactor. In this reactor the solution decomposes to ammonia gas, CO_2 and water vapor. A pressure control valve then regulates the supply of the gas mixture to the SCR unit.

The treated offgas stream is then sent to the caustic scrubber for iodine and sulfur dioxide removal and for final cooling of the offgas stream. At this point the offgas is about 230°C.

Caustic Scrubber

The caustic scrubber removes molecular iodine and sulfur dioxide and provides cooling. The offgas stream enters the bottom side of the scrubber and flows upward through a packed bed. The scrubbing liquid is introduced through a distributor at the top of the packed section of the column and trickles downward through the packing media. To neutralize the collected acid gases, a 5 molar sodium hydroxide solution is added periodically to the caustic scrubber bottoms vessel. This maintains the scrub solution at around 0.5 molar NaOH. The offgas is then discharged through a mist eliminator. The offgas enters the caustic scrubber about 190°C with a 34% relative humidity. Through the scrubber the offgas is cooled to around 70°C and exits at nearly 100% relative humidity. The scrubbing liquid drains into the caustic scrubber bottoms vessel and is recirculated

Carbon dioxide generated in the melter and the catalytic oxidation unit is also removed in this scrubber. Carbon dioxide is neutralized to sodium bicarbonate. ^{129}I is removed in this column if it is present either in the form of HI or I_2. The organic portion of organic iodine is removed in the catalytic oxidation unit. The resulting molecular iodine is removed in the caustic scrubber.

After the caustic scrubber, the offgas is released to the environment via the facility stack. At this point the offgas is at 61°C and nearly 100% relative humidity.

References

1. DOE, Waste Treatment Plant, Solicitation No. DE-RP27-00RV14136, 7-31-00 (DRAFT), //www.hanford.gov/orp/procure/solicitations/14136/ draftsolicitation.htm, 2000.

2. Shelley, S., "Controlling Emissions From Fuel and Waste Combustion", *Chemical Engineering,* **106-1**, pg 82ff, (1999).